Center for Understanding and Control of Acid Gas-Induced Evolution of Materials for Energy (UNCAGE-ME)
EFRC Director: Krista S. Walton
Lead Institution: Georgia Institute of Technology
Start Date: August 2014

***Mission Statement**: To develop a deep knowledge base in the characterization, prediction, and control of acid-gas interactions with a broad class of materials to accelerate materials discovery for large-scale energy applications.*

Acid gases are ubiquitous in multiple large-scale energy applications. For example, SO_x and NO_x are critical components of energy-related gas mixtures. Modern flue gas management involves catalysts that convert these species to less harmful products, as well as membrane and sorbents that can be degraded by these gases. As illustrated in Figure 1, degradation and deactivation effects of acid exposure can reduce access to active metal sites, block pores, reduce overall porosity, and destroy favorable textural properties. These degradation effects are often decisive factors in the practical use of materials such as sorbents for carbon capture, acid gas conversion, and natural gas purification. There are two major challenges in solving these problems: the mechanism of acid gas interactions with complex materials is poorly understood, and the ability to predict or prolong the lifetimes of functional materials is severely limited.

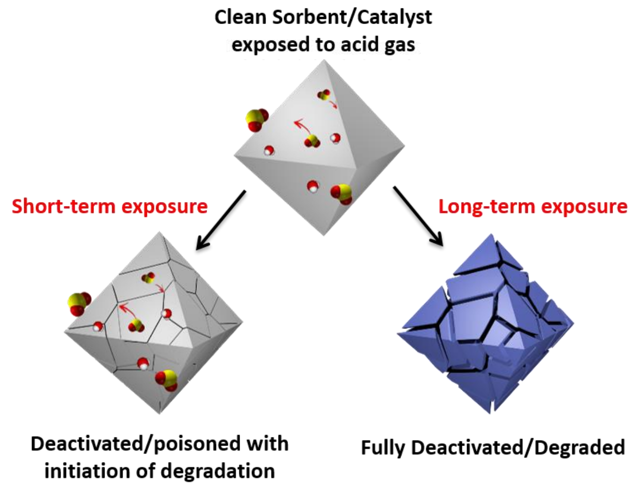

Figure 1: Changes induced in sorbents/catalysts upon short- and long-term exposure to acid gases. Materials are typically treated as passive in these environments, but can change dramatically upon exposure to SO_x, NO_x, and H_2S.

Objectives

UNCAGE-ME seeks to provide a fundamental understanding of acid gas interactions with solid materials through integrated studies of the interaction of key acid gases (CO_2, NO_2, NO, SO_2, H_2S) with a broad range of materials. We combine the application of *in situ* molecular spectroscopic studies of both the surface functionalities and bulk structures of materials relevant to catalysis and separations under relevant environmental conditions with complimentary multiscale computational and theoretical modeling of acid gas interactions with solid matter. Insights gained by the multi-investigator, multidisciplinary teams will allow us to achieve the following long-term, 4-Year Goals set forth for the Center:

1. Develop a deep knowledge base characterizing acid gas interactions applicable to a broad class of materials.
2. Develop fundamental knowledge allowing practical predictions of materials interacting with complex gas environments on long time scales.
3. Advance fundamental understanding of the characterization and control of defects in porous sorbents.
4. Accelerate materials discovery for large-scale energy applications by establishing broadly applicable strategies to extend material stability and lifetime in the presence of acid gases.

Center Research Team and Scientific Organization

The Center conducts research in three major thrust areas:

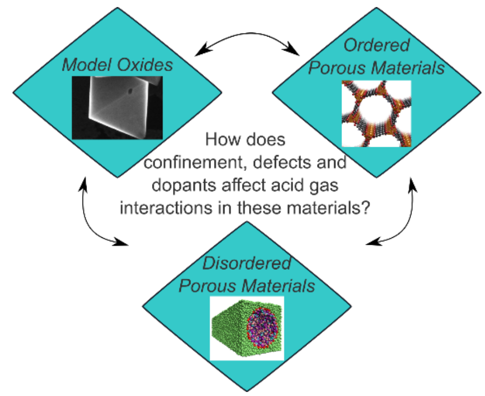

Figure 2: Overview of three research thrust areas.

Thrust I - Model Metal Oxides: The interaction of small amounts of acid gases with sorbents/catalysts can dramatically affect their surface chemistry, useful lifetime and ease of regenerability. The overarching goal of Thrust I is to gain a molecular level understanding of the surface chemistry of acid gas interaction with model sorbents (oxides) and catalysts (supported metals and oxides) and their structural evolution. This will provide structure-performance relationships for the rational design of more efficient sorbents for acid gas removal and more stable catalysts for CO_2 utilization.

Thrust II - Ordered Porous Materials: The stability of crystalline, high surface area adsorbents such as metal-organic frameworks (MOFs) towards acid gases is a major obstacle in their development as successful materials for energy-related applications. The main goal of this Thrust is to determine the structural features that control this (in)stability. The main hypothesis is that acid gas stability of such materials, and their potential degradation upon exposure to acid gases, are driven by local defects or centers susceptible to acid attack distributed throughout the material. If this is the case, characterization and control of the local defects/reactive centers is vital to enhancing material performance.

Thrust III - Disordered Porous Materials: Our efforts using crystallographically ordered porous materials in **Thrust II** will be complemented by **Thrust III**, which will center on disordered porous materials including templated or carbide-derived carbons and supported amines. Both classes of materials have significant heterogeneity with regard to structure in the final form used in acid gas separations and catalysis, yet both are made from more well-defined precursors or supports, which gives a basis for understanding the complex structures of these materials. **Thrust III** aims to create a body of knowledge that ultimately relates the support structure and amines/heteroatoms/defects to its interaction, stability and/or degradation characteristics in the presence of NO_x, SO_x, CO_2, and H_2O.

Center for Understanding and Control of Acid Gas-Induced Evolution of Materials for Energy (UNCAGE-ME)	
Georgia Institute of Technology	Krista Walton, Christopher Jones, David Sholl, Sankar Nair, Michael Filler, Phillip First, Ryan Lively, Thomas Orlando
Oak Ridge National Laboratory	Sheng Dai, Gernot Rother, Tjerk Straatsma, Bobby Sumpter, Zili Wu
University of Alabama	David Dixon
University of Florida	Sergey Vasenkov
Lehigh University	Jonas Baltrusaitis, Israel Wachs
Pennsylvania State University	Susan Sinnott
Washington University St. Louis	Sophia Hayes
University of Wisconsin	JR Schmidt

Contact: Krista Walton, UNCAGE-ME Director, walton@efrc.gatech.edu
404-894-5254, http://efrc.gatech.edu